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Overcoming Hydrogen Losses in Fuel Cells: A Membrane-based Approach to Sustainable Energy

Hossein Pourrahmani, Timur Ashirov, and Ali Coskun§*

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Abstract: Hydrogen (H_2) is increasingly recognized as a key candidate to replace fossil fuels due to its high energy density, zero-carbon combustion, and compatibility with fuel cell technologies. Fuel cells offer an efficient means to convert hydrogen into electricity, with only water as a byproduct, making them a cornerstone for the energy transition. However, challenges remain in the widespread adoption of hydrogen, including production methods (green, blue, and grey hydrogen), transportation, and associated losses during fuel cell operation. A critical issue is hydrogen purge losses, where unreacted H_2 is vented to maintain fuel cell efficiency and durability. This article explores the fundamentals of H_2 fuel cells, purge losses, and the environmental implications. Potential solutions are examined, such as catalytic burning and recirculation systems, to minimize the hydrogen losses in fuel cell strategies. An innovative hydrogen recovery membrane, the SEPARATIC-H_2, developed at the University of Fribourg, has been showcased to enhance fuel cell efficiency while reducing H_2 waste. By addressing these challenges, hydrogen can reach its potential, accelerating the transition toward a sustainable, low-carbon future.

Keywords: Efficiency · Fuel cell · H_2 waste reduction · SEPARATIC-H_2

Dr. Hossein Pourrahmani is a distinguished scientist in mechanical and energy engineering, focusing on computational science, artificial intelligence, and advanced energy systems. He holds a PhD in Energy from EPFL and has significantly advanced renewable energy technologies, particularly in proton exchange membrane fuel cells and electrolyzers. He authored '*The decentralized hydrogen revolution using artificial intelligence, internet of things, and blockchain*', highlighting AI's role in the hydrogen economy. He has an outstanding publication record and collaborations with organizations like the Department of Energy and the European Commission.

Dr. Timur Ahirov was born in Tajikistan and obtained his BSc from the Department of Middle East Technical University, Turkey. Later, he joined the group of Prof. Ali Coskun at the University of Fribourg, Switzerland, for his PhD studies (2018). Timur received the prestigious 2022 Swiss Nanotechnology PhD award for his thesis. He also received the 2023 ACS Global Outstanding Graduate Student Award in Polymer Science and Engineering and the 2023 Vigener Prize. He received multiple grants including the bridge proof-of-concept PoC, GRS InnoBooster, Innosuisse and PBS Implementation grant to scale up and commercialize membranes for CO_2 capture from air and flue gas, hydrogen recovery, and to establish his startup.

Prof. Ali Coskun received his PhD degree in chemistry from Middle East Technical University, Ankara, Turkey. He then joined the laboratory of Prof. J. Fraser Stoddart as a postdoctoral research associate at Northwestern University, where he developed dynamic metal-organic frameworks and artificial molecular machines. He started his independent career at the Korea Advanced Institute of Science in Technology in 2012. In 2017, he moved to the University of Fribourg, Switzerland, as a professor. He is currently developing porous organic polymers for CO_2 capture, separation, and conversion, heterogeneous catalysis, environmental remediation, and high-energy density Li-ion batteries. His research efforts have been recognized with the Swiss Green and Sustainable Chemistry Award 2023.

1. Introduction

Hydrogen (H_2) is gaining significant attention as a clean energy carrier capable of replacing fossil fuels in various applications, particularly through fuel cell technologies in mobility,[1] maritime,[2] and aerospace applications.[3] In recent years, the commercialization of hydrogen fuel cell technology has accelerated, with major industry players investing in its development.[4] Companies such as Ballard Power Systems,[5] Plug Power,[6] and Toyota[7] lead the deployment of fuel cell electric vehicles (FCEVs), while ABB,[8] H_2 Green Ships and Zero Emission Industries drive innovation in hydrogen-powered maritime solutions.[9] In the aerospace sector, companies like Doosan Mobility Innovation, Intelligent Energy, and H3 Dynamics are using hydrogen fuel cells for drones and unmanned aerial vehicles (UAVs), enabling longer flight durations compared to conventional lithium-ion batteries.[10] Proton Exchange Membrane Fuel Cells (PEMFCs), Solid Oxide Fuel Cells (SOFCs), and Alkaline Fuel Cells (AFCs) efficiently convert hydrogen into electricity while emitting only water as a

*Correspondence: Prof. A. Coskun, E-mail: ali.coskun@unifr.ch
Department of Chemistry, University of Fribourg, Fribourg, CH-1700, Switzerland

byproduct.[11] Toyota's Mirai,[7] Hyundai's Nexo,[8] and Honda's Clarity[12] have demonstrated the viability of hydrogen-powered passenger vehicles, while heavy-duty transport applications such as trucks, buses, and trains are being developed by Daimler and Alstom.[13] In the maritime industry, green hydrogen-powered vessels are being designed to comply with International Maritime Organization (IMO) regulations, reducing reliance on heavy fuel oil (HFO) and decreasing sulfur and carbon dioxide emissions.[14] Meanwhile, in aerospace applications, hydrogen fuel cells are being explored for drones, regional aircraft, and even commercial aviation as a way to decarbonize air travel.[15]

However, the adoption of hydrogen fuel cells still faces significant technical challenges, including hydrogen storage and distribution.[16] Hydrogen has the highest gravimetric energy density of any fuel (120 MJ/kg), but its low volumetric energy density creates significant challenges. Accordingly, hydrogen must be stored either at high pressures (350–700 bar) or at cryogenic temperatures, increasing complexity and cost (Fig. 1). Addressing these challenges is critical to ensuring the long-term viability of hydrogen fuel cells as a clean energy solution.[17]

The lifetime of fuel cells is also limited by issues such as membrane degradation, catalyst poisoning, and water management problems.[18] Current fuel cell designs require periodic purging to maintain performance, leading to economic inefficiencies and potential emission concerns. Although periodic purging is necessary to ensure durability and prevent the build-up of contaminants such as nitrogen and water,[19] a portion of valuable hydrogen is inevitably vented, leading to losses. These losses reduce overall efficiency and contribute to indirect greenhouse gas emissions if not managed properly. Hydrogen loss not only affects the economic feasibility of fuel cells but also poses potential environmental and safety concerns. Although hydrogen itself is not a greenhouse gas, its uncontrolled release into the atmosphere can indirectly contribute to global warming by affecting atmospheric chemistry. Recent studies have suggested that increased hydrogen leakage into the atmosphere could alter the oxidative capacity of the atmosphere and impact methane (CH_4) concentrations, a potent greenhouse gas. Since hydrogen reacts with hydroxyl radicals, which play a crucial role in breaking down methane, an increase in atmospheric hydrogen levels could lead to prolonged methane lifetimes, amplifying its global warming potential. Furthermore, hydrogen emissions can affect stratospheric water vapor concentrations, which in turn influence ozone depletion. Although hydrogen fuel cells are promoted as zero-emission technology, these secondary climate effects highlight the importance of addressing hydrogen purge losses as a crucial step for improving a fuel cell systems' sustainability and economic viability. Therefore, implementing hydrogen recovery strategies to minimize purge losses is essential.

1.1 Fuel Cell Types

Hydrogen fuel cells operate based on electrochemical reactions, where hydrogen is oxidized at the anode and oxygen is reduced at the cathode to generate electricity.[20] Fig. 2 shows a schematic of a typical PEMFC, in addition to the balance of plant and the proposed gas separator (SEPARATIC-H_2) for hydrogen recirculation.

Proton exchange membrane fuel cells: In PEMFCs, the membrane, which is usually polymeric, is designed to conduct protons while blocking electrons and reactant gases. However, due to the thinness of the membrane (typically 20–50 µm), a fraction of hydrogen molecules permeate through it and reach the cathode side without participating in the reaction.[21]

Solid oxide fuel cells: SOFCs operate at high temperatures (600–1000 °C) and use a ceramic electrolyte, allowing them to achieve higher efficiencies. However, the hydrogen loss mechanisms in SOFCs are different from those in PEMFCs.[22] Since SOFCs use oxide ion conducting electrolytes, hydrogen loss primarily occurs through unreacted fuel exhaust, which contributes to efficiency losses and must be recirculated or burned. Although SOFC electrolytes, such as Yttria-Stabilized Zirconia (YSZ), are

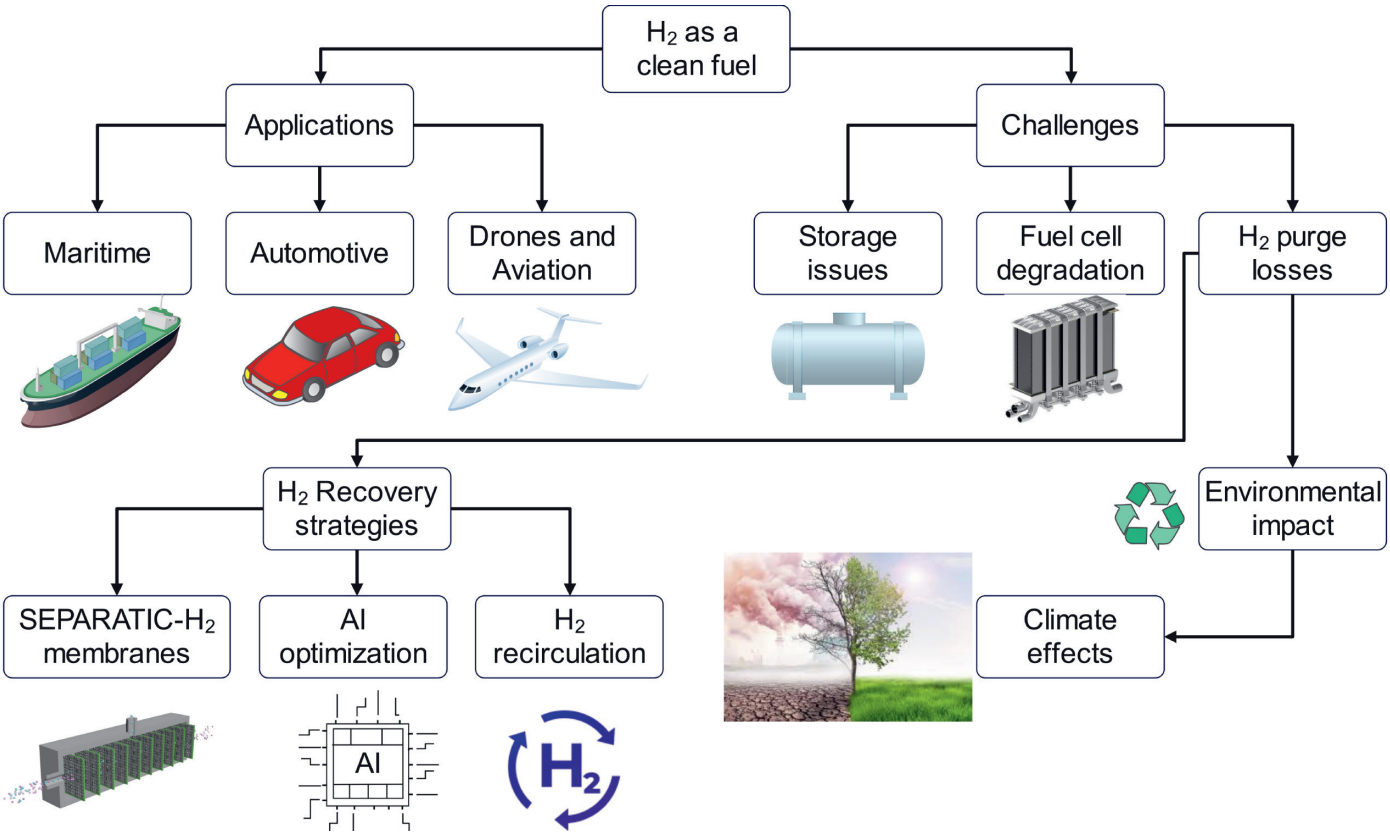

Fig. 1. A schematic flowchart of hydrogen as a fuel: Challenges and possible solutions.

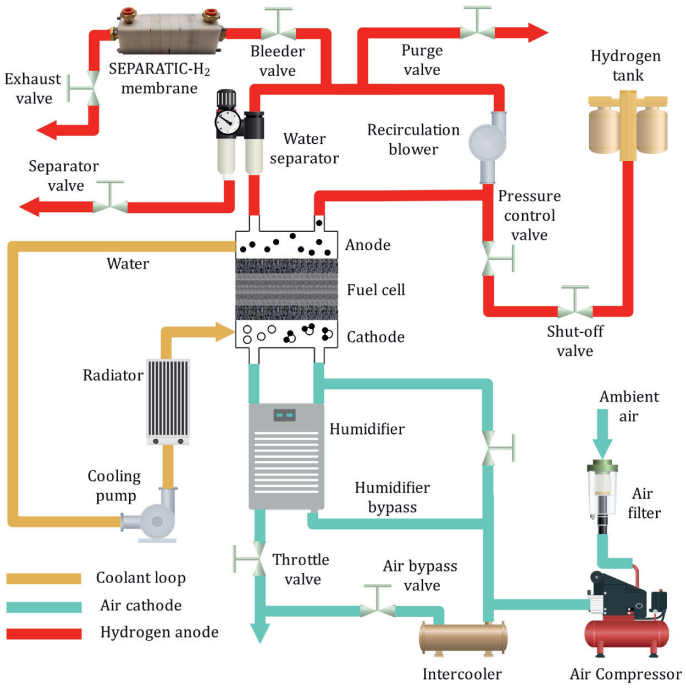

Fig. 2. A schematic of the PEMFC and the balance of plant.

designed to be gas-tight, small leakages can occur due to microcracks or imperfections.

Alkaline fuel cells: AFCs use a liquid electrolyte such as potassium hydroxide (KOH) to operate at moderate temperatures between 60 and 90 °C. Hydrogen losses in AFCs occur due to crossover through the porous electrode, gas solubility, and electrolyte losses.[23] Unlike PEMFCs, AFCs suffer from hydrogen dissolution in the alkaline electrolyte. Dissolved hydrogen eventually escapes from the electrolyte, leading to gradual losses over time. During fuel cell operation, contaminants such as nitrogen (N_2), water vapor (H_2O), and carbon monoxide (CO) accumulate in the anode chamber. These contaminants originate from various sources, such as N_2 ingress due to air crossover through the membrane, and the formation of water vapor as a result of existing electrochemical reactions inside the fuel cell, which can lead to flooding or performance loss.[24] Fuel cell durability is a key concern in commercial applications, as degradation affects performance and longevity.[25] Another possible source of degradation is platinum catalyst poisoning; accumulated impurities bind to the catalyst and inhibit hydrogen oxidation. To mitigate degradation, controlled purge cycles are implemented based on monitoring key parameters such as voltage degradation rate and membrane resistance.

In summary, hydrogen purge losses are an unavoidable aspect of fuel cell operation, but their impact can be minimized through optimization strategies. There is a need for hydrogen separator technologies that can eliminate hydrogen losses in fuel cells. Subsequent sections will explore advanced methods for reducing hydrogen losses and the potential of SEPARATIC-H_2 adsorptive membranes.

2. Hydrogen Capture and Recovery Technologies

Current hydrogen separation technologies are based mainly on adsorbents and membranes; adsorbents have porous structures that can store H_2 within their pores, while membranes have pores small enough to let H_2 pass while larger gas molecules are blocked.

2.1 *Hydrogen Capture and Recovery Using Adsorbents*

Porous materials such as metal-organic frameworks (MOFs),[26] covalent organic frameworks (COFs),[27] and porous organic polymers (POPs)[28] have been extensively studied for hydrogen separation and recovery owing to their large surface areas, tunable structures and pore sizes, and high porosities.[29] For example, NU-100 MOF showed exceptional H_2 uptake of 14.1 wt% at 77 K and 70 bar.[30] Similarly, MOF-210 and MOF-5 also showed the H_2 uptake capacities of 14.3 and 9.7 wt%, respectively.[31] Our research group also studied the potential of MOFs for H_2 storage applications; we blended graphene oxide with HKUST-1 MOFs to achieve the H_2 uptake performance of 1.4 wt% at 77 K and 1.1 bar.[32] In another example, bimetallic MOFs containing palladium (Pd) and copper (Cu) were synthesized, namely the PdCuBTC and PdCuBTB (BTC = benzene-1,3,5-tricarboxylic acid and BTB = 1,3,5-tris(4-carboxyphenyl)benzene), that exhibited H_2 uptake capacities of 2.6 and 2.9 wt% at 67 K and 1.3 bar, respectively.[33] These examples highlight the potential of MOFs for H_2 storage and separation applications.

COFs have been predicted to have exceptional H_2 uptake capacities exceeding the best-performing MOFs, with COF-108's theoretical capacity of 18.9 wt% at 77 K and 100 bar.[27] However, even the best-performing COFs did not surpass MOFs, with COF-102 demonstrating 6.8 wt% H_2 capture capacity.[34] The reason for this is that the presence of metal in the framework enhances the H_2 uptake capability by creating additional binding sites.[33] For instance, it has also been predicted that lithium (Li) doping could enhance the uptake properties of COFs due to the strong affinity of the Li^+ cation to H_2, with Li doped COF-202 predicted to exhibit more than a 3-fold increase in uptake, reaching 7.83 wt% at 77 K and 100 bar.[35]

POPs, on the other hand, have been extensively studied for H_2 storage and separation. Porous aromatic frameworks (PAFs), a subclass of POPs, are known for their exceptional H_2 uptake performance due to their all-carbon structures. PAF-1, with its remarkable surface area of 5600 $m^2\ g^{-1}$, demonstrated 10.7 wt% H_2 uptake at 77 K and 48 bar.[36] Similarly, the porous polymer network PPN-4 exhibited a H_2 capture capacity comparable to MOFs with 13.6 wt% at 77 K and 80 bar.[37] We also demonstrated that porous polyphenylenes composed of all-carbon structures, namely the 2D- and 3D-pPPs, showed 7.4 and 7.1 wt% H_2 uptake performances at 77 K and 109 bar, respectively.[38] Although these studies demonstrate the potential of porous materials for H_2 separation, they are far from commercialization for this purpose. Current research is more focused on H_2 storage using porous materials rather than separation. In addition, the uptake conditions are measured at cryogenic temperatures and high pressures, far from standard conditions, making them unsuitable for H_2 recovery from the fuel cell purge.

2.2 *Hydrogen Separation and Recovery Using Membranes*

Membranes are used extensively for hydrogen separation applications.[39] Commercial polymeric membranes have already been employed for H_2/N_2 separation in various separations, including ammonia and methanol production, and steel making processes, where operating pressures can range between 5–100 bar.[40] One of the challenges associated with membranes is the trade-off between permeance and selectivity; fast-permeating membranes usually have lower selectivities and vice versa.[41] This also means very high pressures are required to drive H_2 through the membrane. There has been significant progress made with the discovery of polymers of intrinsic microporosity (PIM),[42,43] where higher H_2 permeances were obtained. For instance, PIM-1 achieved H_2 permeance of 136 GPU (1 GPU = 3.35×10^{-10} mol s^{-1} m^{-2} Pa^{-1}) and H_2/N_2 selectivity of 8.95.[43] Mixed matrix membranes (MMMs) incorporating fillers such as MOFs and COFs in the polymer matrix have been shown to exhibit improved separation performances.[44] For example, Zhu and coworkers incorporated CAU-21 MOF into the PIM-1 matrix and reached H_2 permeance of 562 GPU and

H_2/N_2 selectivity of 127 at 15.2 wt% loading.[45] Despite the performance improvements, polymeric membranes suffer from aging issues, particularly in humid conditions and at high operational temperatures.[44] This, combined with the requirement of high pressures, renders polymer-based membranes are unsuitable to address the fuel cell purge losses.

The permeances of polymer-based membranes are still too low to achieve reasonable permeation performance at low pressures (<5 bar), which limits their applications on small scales.[40] Novel membranes based on 2D materials like graphene could potentially address this issue with their ultrahigh permeabilities. For instance, exfoliated graphitic carbon nitride (g-C_3N_4) films reached H_2 permeance of 1450 GPU and H_2/N_2 selectivity of 48.[46] Porous graphene membranes can achieve even higher H_2 permeances of up to $6x10^7$ GPU.[47] However, they also suffer from low selectivities due to current pore generation methods. In an attempt to solve this problem, our group studied the gas transport kinetics by tuning the pore size of graphene *via* stepwise gold deposition. In this approach, at a 100 nm gold layer, we reached H_2/CO_2 selectivity of 31.3 with H_2 permeance of $2.23x10^5$ GPU.[48] Potentially 2D membranes can be used to address the fuel cell purge losses if applied correctly; however, the operational durability under humid conditions needs to be validated.

2.3 *Thick Palladium Membranes for Hydrogen Recovery*

Hydrogen gas can permeate through a palladium (Pd) membrane by dissociating into atomic hydrogen on the surface, diffusing through the metal lattice, and then recombining into molecular hydrogen on the opposite side.[49] Accordingly, platinum group metals offer very high H_2 selectivities and have been studied extensively for membrane applications.[50] These membranes are commercialized more for H_2 production applications by companies such as H_2SITE and MTR Corp. The Pd film needs to be free of cracks and defects that can result in selectivity loss. Therefore, current methods of preparation of Pd membranes lead to membranes with thicknesses in the range of several microns, increasing the costs further. However, the H_2 permeation is extremely low at room temperature, and to achieve meaningful permeation rates, Pd membranes operate at high temperatures above 300 °C.[51] This process not only increases the operational costs, but also reduces the lifetime of the membranes significantly due to microcracks and defect formation,[52] with the best-performing examples achieving several hundred hours of operational lifetime. Moreover, the presence of high humidity can shorten the lifetime of the Pd membranes. For instance, Basile and coworkers coated an 8 µm thick Pd-Au alloy (12 wt%) on an alumina ceramic membrane and analyzed H_2 permeation at 400 °C, and observed H_2/N_2 selectivities up to 500 along with a permeance of 8160 GPU.[53] However, the membrane performance significantly diminished after 600 h of operation. As can be seen, thick Pd membranes have great potential for solving the issue of H_2 loss in fuel cells; however, high production and operational costs, and relatively short lifetime make their integration challenging.

2.4 *Other Hydrogen Recovery Methods*

Besides membranes and adsorbents, H_2 can be recovered by using electrochemical separation, where H_2 gas is passed through a membrane where it is selectively converted into H^+ with an electrical current. In a nutshell, it is a modified electrolyzer that separates H_2 from other gases.[54] Such systems are already being commercialized by companies such as Skyre Inc. and are particularly useful for hydrogen recovery from dilute hydrogen streams, offering the advantage of high selectivity and low energy consumption. However, small-scale implementation of such separation processes is challenging due to the high cost of production.

2.5 *The Proposed Solution*

As summarized above, currently, there is no active solution for eliminating H_2 purge losses in fuel cells. It is possible to minimize these losses by optimizing the fuel cell operation and the addition of a recirculation pump, which recycles unburnt fuel to lower the H_2 concentration before purge; however, it cannot eliminate H_2 loss. To prevent indirect environmental consequences of H_2 emissions, another alternative is to oxidize it using a catalytic burner. However, this results in a loss of valuable hydrogen and jeopardizes the safety of the fuel cell systems.[55]

To address specifically the H_2 purge losses, we at the University of Fribourg have developed a novel, patent-pending, adsorptive separation membrane technology (SEPARATIC-H_2) that can selectively capture H_2 from the mixture and put it back into the fuel cells. The unique design of combining an adsorbent with the membrane makes it possible to remove any gas from the mixture with high selectivity; the selected gas sticks to the adsorbent while non-interacting gases pass through the membrane. The adsorbed gas can be released using a combination of vacuum and heat. For H_2 separation, we used Pd as an adsorbent and achieved a 100% capture rate from a mixture of helium and hydrogen, with no negligible loss after 20 cycles.[56] This process offers high selectivities and high permeation rates at the same time, and is suitable under the harsh operational conditions required for fuel cells. Current efforts are focused on scaling this technology, testing, and integrating it into the fuel cell systems. The hydrogen recovery process using Pd membranes operates in the two main stages of absorption and diffusion. Integrating the SEPARATIC-H_2 membranes into the fuel cell systems offers multiple benefits (see Fig. 3), including:

- Prevention of H_2 loss by selectively recovering and recycling hydrogen; Pd-based membranes help minimize purge losses.
- Improved fuel cell efficiency; hydrogen recovery reduces the need for continuous hydrogen supply, reducing overall fuel consumption.
- Improved fuel cell safety; removing H_2 emissions will also prevent uncontrolled reactions that can lead to explosions.
- More sustainable fuel cells; preventing H_2 loss will also prevent potential environmental consequences of H_2 emissions.
- Low operational costs; SEPARATIC-H_2 membranes require low energy to operate, compared to thick Pd membranes.

Given the growing demand for hydrogen fuel cells and the urgency to address hydrogen purge losses, we explored the fundamental mechanisms behind hydrogen purge losses in fuel cells, the environmental and economic impacts of hydrogen emissions, the role of leading companies in the commercialization of hydrogen-powered transportation, and state-of-the-art hydro-

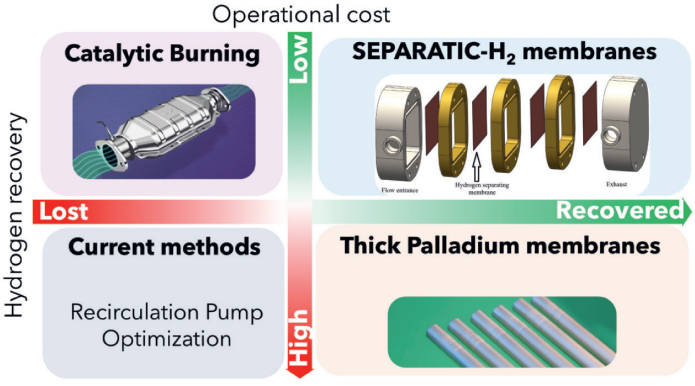

Fig. 3. Illustration of the SEPARATIC-H_2 separator in comparison to other potential technologies in terms of operational costs and hydrogen recovery.

gen recovery solutions, including Pd-based membranes. From a regulatory perspective, the absence of strict policies governing hydrogen emissions and recovery strategies remains a challenge. A global policy framework incentivizing hydrogen capture and recycling could drive industrial adoption of advanced purge management techniques. Additionally, a Life Cycle Assessment (LCA) of hydrogen recovery solutions can provide an in-depth understanding of their environmental and economic trade-offs, ensuring that proposed solutions align with sustainability goals. Standardizing best practices for hydrogen management in fuel cells will be essential to maximizing the benefits of hydrogen as a clean energy carrier.

3. Conclusions

In this article, we studied a comprehensive analysis of hydrogen purge losses, their causes, and their environmental and economic implications. Different types of fuel cells were discussed, including PEMFCs, SOFCs, and AFCs, each of which experiences hydrogen loss through various mechanisms. Understanding these loss pathways is crucial in developing effective mitigation strategies. Among the solutions explored, Pd-based membranes have emerged as a promising technology for hydrogen recovery due to their high selectivity and permeability. Additionally, other advanced separation techniques, including porous materials, electrochemical hydrogen separation, and polymer-based membranes, offer potential pathways for enhancing hydrogen retention. While these technologies present opportunities for significant efficiency improvements, challenges remain in terms of scalability, cost, and durability, which require further research and development.

Beyond technological advancements, policy and regulatory frameworks play a crucial role in minimizing hydrogen losses and promoting efficient recovery strategies. A lack of strict regulations on H_2 purge management has led to inefficiencies in fuel cell operation, highlighting the need for international standards and incentive mechanisms. Implementing LCA and AI-driven hydrogen management systems can optimize purge cycles, reducing waste while maintaining system longevity. Additionally, financial incentives such as credit schemes and government subsidies can encourage industry-wide adoption of hydrogen recovery technologies. By integrating cutting-edge recovery technologies, such as the SEPARATIC-H_2, refining operational strategies, and establishing supportive policies, hydrogen fuel cells can reach their full potential as a cornerstone of clean energy systems. Future research will be focused on improving membrane durability and optimizing hydrogen recirculation systems. With continued advancements and collaborative efforts across industry, academia, and policymakers, hydrogen can become a truly sustainable and economically viable energy carrier, accelerating global decarbonization efforts.

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